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THE ABSORPTION SPECTRUM OF CALCIUM VAPOR: 1660 - 2028 Å

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By

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Abstract

The vacuum ultraviolet absorption spectrum of calcium vapor was observed in an attempt to discover transitions to new energy levels. Seven new levels have been found between 51000 and 59000 cm^{-1}, three of which are ascribed to the 3d5p, 3d6p, and 5s4p configurations, with the remaining four lines resulting from transitions to 3d4f and 3d5f configurations. The 3d5p $^3P^o_1$ level reported by Ditchburn and Hudson at 52225 cm^{-1} is now found at 51909 cm^{-1}. A graph of relative cross-section for photoionization from the ground state of calcium is shown between 1660 Å and the principal series limit. A shock tube was used to observe several transitions to autoionizing levels from excited states.

1. Introduction

A recent paper by Garton and Codling (1965) reported many new energy levels of Ca I above the principal series limit, although several levels expected between 51000 cm^{-1} and 59500 cm^{-1} were not found. Also, since the $4s^2\ ^1S_0 - 3d5p\ ^3P_1^\circ$ transition reported at 1914.8 Å by Ditchburn and Hudson (1960) has not been confirmed by other observers (Kaiser 1960; Garton and Codling 1965), doubts have arisen about the location of this level. The present investigation was therefore undertaken in an attempt to observe transitions to the missing levels and to determine the location of the $3d5p\ ^3P_1^\circ$ level. A photoelectric scanning monochromator was used to measure the photoionization cross-section of calcium from 1660 Å to the series limit. Seven new lines were discovered and the classification of all lines observed in this spectral region was attempted. Oscillator strengths of seven lines were measured relative to the series limit cross-section. The transition to the $3d5p\ ^3P_1^\circ$ level has been found to be 11.7 Å to the red of the position of Ditchburn and Hudson.

2. Experimental

A Burrell Electric Furnace, Model BT-1-23, contained a 50-cm stainless steel test section sealed at each end by quartz lenses, selected for high transmission at vacuum ultraviolet wavelengths. Stops were placed at each end and in the middle of the test section and an examination of the tube after the experiment showed that no calcium had condensed on the wall outside the test section.

Since the current to the furnace was adjusted in discrete steps, the desired temperatures often could not be held constant and variations of $\pm 5°C$ were typical during many absorption scans. A thermocouple mounted near the outside of the test section at the middle of the furnace monitored the furnace temperature, so that temperature variations were noted on all scans.

Before the absorption scans of calcium vapor were taken, the empty test section was outgassed by heating it to 1000°C while helium at a pressure of 10 torr flowed through the tube. The tube was then cooled sufficiently to allow two stainless steel boats containing purified calcium turnings to be introduced near each end of the test section. A flow of helium was maintained through the tube while the vacuum was broken to minimize contamination with air. The tube was then sealed with the quartz lenses and a helium flow at about 10 torr was again maintained to sweep out contaminants which vaporized as the furnace was being heated.

At a temperature of about 500°C, the flow of helium was stopped and a helium pressure of about 100 torr was maintained to slow the diffusion of calcium atoms. With the helium flow stopped, many broad absorption bands were observed with a half-width of about 12 Å, presumably due to molecular contaminants in the tube. The absorption bands rapidly disappeared when the furnace was heated to 900° or 1000°C. After the disappearance of these bands, the tube was cooled and the background continuum was scanned; scans were then made of the calcium absorption at various temperatures. Finally the tube was cooled and the continuum recorded.

A hydrogen positive-column discharge, operated at 400 milliamps and 700 microns pressure, provided the background continuum. Emission bands of CO were visible when the discharge was first turned on, but these disappeared during the one-hour discharge warm-up. The quartz lens which sealed the test section also served to confine the discharge.

The hydrogen discharge was focused on the entrance slit of a one-meter McPherson scanning monochromator, with an automatic focus normal-incidence mount. The gold-coated grating, 51 x 94 mm, was ruled with 30,000 lines/inch, which gave a reciprocal dispersion of 8.5 Å/mm. To measure the slit widths, the labeled widths were assumed to be accurate except for a zero-point error. The photomultiplier output was then measured as a function of labeled slit width. The intercept of the resulting linear function gave the zero-point error. The derived slit widths were 7 and 13μ respectively. The lines of the principal series could be seen to n = 40, so that two weak lines 0.15 Å apart would appear with separate peaks.

An EMI 6256 S/A photomultiplier tube, selected for low noise, was mounted behind the exit slit. At a voltage of 1250V, the measured dark current was 2.1×10^{-9} amp. A Keithley 600 A electrometer measured the anode current, and the voltage output of the electrometer was displayed on a Speedomax chart recorder at a speed of 8 inches per minute. The relevant data about the spectral scans are summarized in Table 1.

Since the continuum intensity generally decreased at shorter wavelengths, gain changes were required on the electrometer to yield useful deflections over the desired wavelength range. The chart deflections for zero signal, dark current, and scattered light were measured periodically at all gain settings, with the scattered light signal derived from the deflection at the center of a saturated absorption line or at a wavelength to the violet of the quartz transmission limit.

3. Data Reduction

A wavelength scale was marked on the spectral scans, with the Mg I principal series and the C I emission line at 1930.9 Å serving as standards. Measurements were normally made every 2.5 Å, but points were measured at shorter intervals when the cross-section varied rapidly with wavelength. All scans were reduced to digital form by measuring the deflections relative to the scattered light background for all wavelengths, except in regions where saturation made useful measurements impossible.

Traces 22, 23, 32, and 33 were used to represent the continuum. Only trace 23 showed any noticeable calcium absorption above the principal series limit; points on this scan in the region of the lines at 1885.9 and 1765.1 Å were therefore rejected. Note that scan 30 showed a generally lower thermocouple temperature than did scan 23, yet considerably more calcium absorption is visible on scan 30, the obvious explanation being that the thermal lag of the test section prevented it from responding quickly to the rapid temperature changes on these scans.

The discharge had obviously brightened considerably during the course of the experiment. The brightness of each scan relative to trace 33 was computed for a broad wavelength region and the values were found to be 0.748, 0.752, and 0.986 for scans 22, 23, and 32 respectively. Each point on these three scans was divided by the appropriate brightness factor and the resulting normalized continua showed the same distribution of brightness with respect to wavelength. Hence the increased brightness was assumed to be independent of wavelength and a function only of time. The intensities of the four continua normalized to trace 33 were averaged at each wavelength to give the best distribution of continuum brightness.

The optical depth at a given wavelength in the furnace is given by the expression

$$\tau_\lambda = \ln\left(\frac{I_\circ}{I}\right) = N \cdot \sigma_\lambda \ ,$$

where $I_\circ$ is the continuum intensity at wavelength λ, I is the residual intensity, N is the number of absorbing atoms in the line of sight, and σ_λ is the atomic cross-section. N was derived from the measured value of τ_λ at the principal series limit, a value of 9.0×10^{-19} cm^2 being used for σ_λ.*

For scans 29 and 30, the series limit absorption was too weak to yield an accurate value of τ_λ, and scan 27 showed very strong absorption at this wavelength. In the region from 1998 to 1953 Å, however, both scans 26 and 27 showed measurable absorption, and the ratio of optical depths for this wavelength interval, 1.46, is also the ratio of number of atoms in the line of sight. Similarly, the value of N for scan 29 was normalized against that for scan 28 in the wavelength ranges 1898 - 1893 Å, while scan 30 was normalized against scan 29 in the region 1905 - 1855 Å. The peak absorption of the lines at 1885.9 and 1765.1 Å must be regarded as less accurately known than the weaker absorption at adjacent wavelengths because this method introduces somewhat greater uncertainty in number density at low temperatures.

The results of scans taken at different temperatures were graphed to give the number of atoms in the line of sight as a function of thermocouple temperature; corrections resulting from temperature changes during the course of a single scan were taken from this graph (Fig. 1).

All optical depths were converted to cross-sections and the points were plotted as a function of wavelength. Since each scan was made at a different temperature with a different correction factor for continuum brightness, the dispersion of points at a given wavelength can be used to estimate the internal accuracy of the cross-sections. The standard deviation in megabarns for the cross-section at a given wavelength is given

*This value is double that given by Ditchburn and Hudson (1960), owing to improved measurement (JANEF 1962) of the dependence of vapor pressure on temperature (Hudson, private communication).

roughly by 0.01 + 0.05 x σ, where σ is the cross-section in megabarns. Each point was weighted before a final mean was taken, with points having a residual intensity between 20% and 80% of the continuum weighted more heavily. The weighted mean cross-sections are shown in Figure 2. The regions marked with a heavy line were not adequately resolved by the monochromator and cannot be considered as accurate.

4. Results and Discussion

The line at 1914.8 Å reported by Ditchburn and Hudson was not visible on the spectral scans, but a previously unknown diffuse line was observed at 1926.5 Å (Fig. 2). The line was also found on a plate (Plate 1) taken by W. H. Parkinson and E. M. Reeves at Imperial College with a three-meter vacuum spectrograph, and on a spectrum of shock-excited CaS taken by the author at Harvard College Observatory with a one-meter vacuum spectrograph. The presence of this line on the spectra of these three independent calcium experiments should establish beyond doubt that a line exists at 1926.5 Å, and the $4s^2\ ^1S_0$ - $3d5p\ ^3P^\circ_1$ identification is highly probable.

The weak line at 1786.28 Å was previously identified as a transition to the $3d6p\ ^3D^\circ_1$ level (Garton and Codling 1965), which requires this level to be strongly perturbed. From the quantum defects of the other members of this series (Fig. 3), this line would be expected at 1769.0 Å, only 0.1 Å away from a sharp line previously ascribed by Garton and Codling to the $3d6p\ ^3P^\circ_1$ level, and the $3d6p\ ^3D^\circ_1$ identification is therefore proposed for this level. A new diffuse feature at 1777.6 Å has the same appearance as the $4s^2\ ^1S_0$ - $3d5p\ ^3P^\circ_1$ line at 1926.5 Å, which suggests the identification of the line at 1777.6 Å as the n = 6 member of the $3dnp\ ^3P^\circ_1$ series. The quantum defects which result from this identification show both the $3d6p\ ^1P^\circ_1$

and $^3P^\circ_1$ levels highly perturbed by about the same amount (Fig. 3). Garton and Codling suggest that the 5s4p $^1P^\circ_1$ level is responsible for the perturbation of the 3d6p $^1P^\circ_1$ level, and a logical extension indicates that the 5s4p $^3P^\circ_1$ has a similar effect on the 3d6p $^3P^\circ_1$, while the 3d6p $^3D^\circ_1$ level remains unperturbed.

Garton and Codling identify the absorption line at 1740.2 Å as possibly the transition to the 5s4p $^1P^\circ_1$ level. However, our spectral scans show a very broad absorption feature of about 15 Å half-width centered about 1724.8 Å, which is more likely to be the $4s^2\ ^1S_0$ - 5s4p $^1P^\circ_1$ line. The line at 1740.2 Å is then ascribed to the 5s4p $^3P^\circ_1$ level. The width of the 1725 Å line is about that expected for the 3d6p $^1P^\circ_1$ level on the basis of the appearance of the transitions to the 3d5p $^1P^\circ_1$ and 3d7p $^1P^\circ_1$ levels, and the wavelength is about as close to the expected value as the 1765.1 Å line; this fact suggests that the 3d6p $^1P^\circ_1$ designation should be applied to the new line at 1725 Å. However, a strong intensity perturbation without a strong perturbation of half-width would be required to explain the relative weakness of the feature at 1725 Å, so that the 5s4p $^1P^\circ_1$ classification is preferable for this line.

The 3dnf $^1P^\circ_1$ and $^3D^\circ_1$ series have been reported by Garton and Codling (1965) for $n > 5$, with the 3dnf $^3P^\circ_1$ series given for $n > 6$. An extrapolation of the quantum defects for the 3dnf $^1P^\circ_1$ and $^3D^\circ_1$ series to the $n = 4$ and $n = 5$ levels yields four wavelengths, all of which agree with observed lines. The $n = 5$ levels at 1709.4 and 1711.5 Å are both somewhat diffuse, in contrast to other members of these series. The 3d5f $^3P^\circ_1$ level was not found, possibly because the weak line expected would not be easily seen if diffuse. If the 1711.5 Å line is attributed to the ^3P, the effective quantum number is 4.889, well below the interpolated value of 4.918 (Fig. 4).

In the region of the 3d4f lines, four sharp lines are visible, although one (1785.3 Å) is very weak. If the remaining lines at 1786.28, 1787.42, and 1788.82 Å are ascribed to the 3d4f $^1P^\circ_1$, $^3P^\circ_1$, and $^3D^\circ_1$ levels, the quantum defects appear plausible (Fig. 4). If the identifications for the 3d4f $^3D^\circ_1$ and $^3P^\circ_1$ levels are interchanged, the relative intensities are in better agreement with the general behavior for higher series members, but the quantum defect for the 3d4f $^3D^\circ_1$ level is then required to be greater than that of the 3d4f $^1P^\circ_1$. A consideration of possible nearby perturbing levels does not indicate that the 3d4f levels should be perturbed enough for such a crossing to occur, so that the initial identification above is proposed as the most satisfactory. The presence of the fourth line is bothersome, although three other weak unidentified lines at 2004.6, 1999.6, and 1996.0 Å are visible which do not fit any possible calcium series and do not agree with any previously observed lines of the known impurities in the furnace, magnesium and barium. These four lines may result from unknown impurities in the furnace. None of these four lines appear on Plate 1, but one would not expect to see lines of this strength on that plate.

The smooth behavior of the quantum defects for the 3dnf levels for the smaller values of n indicates that the wavelengths of the unobserved transitions to the 3d5f $^3P^\circ_1$ and 3d6f $^3P^\circ_1$ levels can be predicted with reasonably high accuracy. The estimated quantum defects of these levels (open circles in Fig. 4) correspond to wavelengths at 1709.9 and 1670.6 Å.

A comparison of the graph of cross-section (Fig. 2) with a similar graph by Ditchburn and Hudson (1960) reveals several major differences. For example, the Ditchburn and Hudson curve gives a cross-section of ≥ 0.3 Mb for all wavelengths except the "window" between 1885.9 Å line and the series limit, while Figure 2 decreases to 0.18 Mb at 1785 Å and 0.05 Mb at 1706 Å. Ditchburn and Hudson measure the peak cross-section for the transitions to the 3d5p $^1P_1^\circ$ and 3d6p $^1P_1^\circ$ levels as 30 and 70 Mb respectively, while the photoelectric measures give 14 and 21 Mb. These differences presumably result from the fact that Ditchburn and Hudson used a fluorite prism spectrograph and photographic film to measure absolute cross-sections, while the present experiment used a grating monochromator and photoelectric detector to measure relative cross-sections.

The scanning monochromator gives better resolution than the fluorite prism spectrograph, but if the peak cross-section values of Ditchburn and Hudson for the lines at 1886 and 1765 Å were seriously affected by resolution, the values should be smaller, rather than larger, than the results derived from the monochromator. Hence the difference in resolving power for the two experiments does not explain the discrepancy. The use of photoelectric detection yields a greater dynamic range and avoids errors which arise in calibration through the use of a characteristic curve. The photoelectric values would therefore be expected to be more accurate.

The very large range in cross-section of calcium vapor requires that the spectrum be recorded at many different vapor pressures. If absolute cross-sections are measured, errors in the dependence of vapor pressure on the temperature appear directly as errors in derived cross-section, whereas in the present experiment the relative number of atoms in the line of sight is found from the observed absorption at a given wavelength and is independent of the curve showing vapor pressure as a function of temperature. Hence the relative cross-sections given here should be more accurate than the relative values of Ditchburn and Hudson, although the present method leaves unsolved the more difficult problem of accurately determining the scale factor to yield absolute cross-sections.

The f-values given in Table 2, determined by integrating the cross-section graphed in Figure 2, are normalized against the series limit absorption of 9.0×10^{-19} cm^2. The derived values depend somewhat critically on the assumed background continuous absorption and the limits of integration. For the 1885.9 and 1765.1 Å lines, the background absorption was assumed to be 0.0 and 0.2 Mb respectively, with the integration extending from 1942 to 1789 Å and from 1780.5 to 1750.4 Å. For the remaining lines whose f-values are given, the background absorption was interpolated from the measured absorption adjacent to the line.

The half-widths of several lines have been measured from Figure 2 and are shown in Table 2. The half-width of the line at 1885.9 Å agrees to 1% with the value obtained in a shock tube by Garton et al. (1964), but the half-widths of the lines at 1885.9 and 1765.1 Å are considerably smaller than the values of Ditchburn and Hudson.

Most wavelengths reported in Table 2 were measured with a Zeiss comparator on the plate taken by Parkinson and Reeves at Imperial College and shown in Plate 1, with the Mg I principal series and the resonance line of Hg I at 1849.497 Å being used for comparison. However, the very broad lines at 1885.9, 1765.1, and 1724.8 Å were measured on the photoelectric spectral scans, with the values listed referring to the wavelength of maximum cross-section. The line at 1670.01 Å could not be measured accurately because of its proximity to an emission line in the hydrogen discharge source, and the wavelength of Garton and Codling (1965) is used here. A comparison of the wavelengths reported here with the measurements of Garton and Codling show a disagreement typically of only 0.01 Å for the sharper lines. The wavelengths of the lines at 1885.9, 1765.1, and 1724.8 Å are probably accurate to about 1 Å. All wavelengths of lines observed in the furnace are given in vacuum.

5. Astrophysical Importance

At the temperatures of stellar atmospheres, where excited levels are populated, transitions to broad autoionizing levels can occur at visible wavelengths. In the solar spectrum, one such multiplet has recently been identified (Mitchell and Mohler 1965) and many other weak diffuse absorption features have been observed (Goldberg et al. 1965). The identification of autoionizing energy levels is thus of astrophysical importance since it allows one to determine which of the observed broad absorptions are caused by autoionizing lines.

Since the furnace spectra reveal only transitions to levels which can combine with the $4s^2\ ^1S_0$ ground state (i.e., levels of odd parity and $J = 1$), the majority of levels above the principal series limit are seen only when combining with excited levels, and a high-temperature source is required to populate these excited levels. A shock tube is now being used to discover new lines, which are being classified. Several lines fit the expected wavelengths for transitions from known excited levels to autoionizing configurations seen in the furnace, as shown in Table 3. Definite indentification is not yet possible, since the high pressures in the shock tube cause many lines to be pressure broadened, so that measurements of accurate wavelengths and half-widths are impossible. A low-density source, such as a hollow cathode, may be required before positive identification can be made.

A comparison of the shock tube and solar spectra has been made, with only two diffuse features appearing at the same wavelengths on both spectra. The two absorptions are at 6230 and

6242 Å, with half-widths of about 6 Å each. The absorption coefficient at line center for the shock tube spectrum must be measured before one can determine whether these features have a common origin in both spectra.

The autoionizing series of neutral calcium are obviously in need of a detailed theoretical treatment. The use of LS notation to describe these levels may not be justified, and accurate classification based on this scheme may not be possible. The observations of all autoionizing lines of calcium vapor within 300 Å of the principal series limit are probably now complete, and a theoretical analysis would contribute much towards an understanding of this complex spectral region.

Acknowledgements

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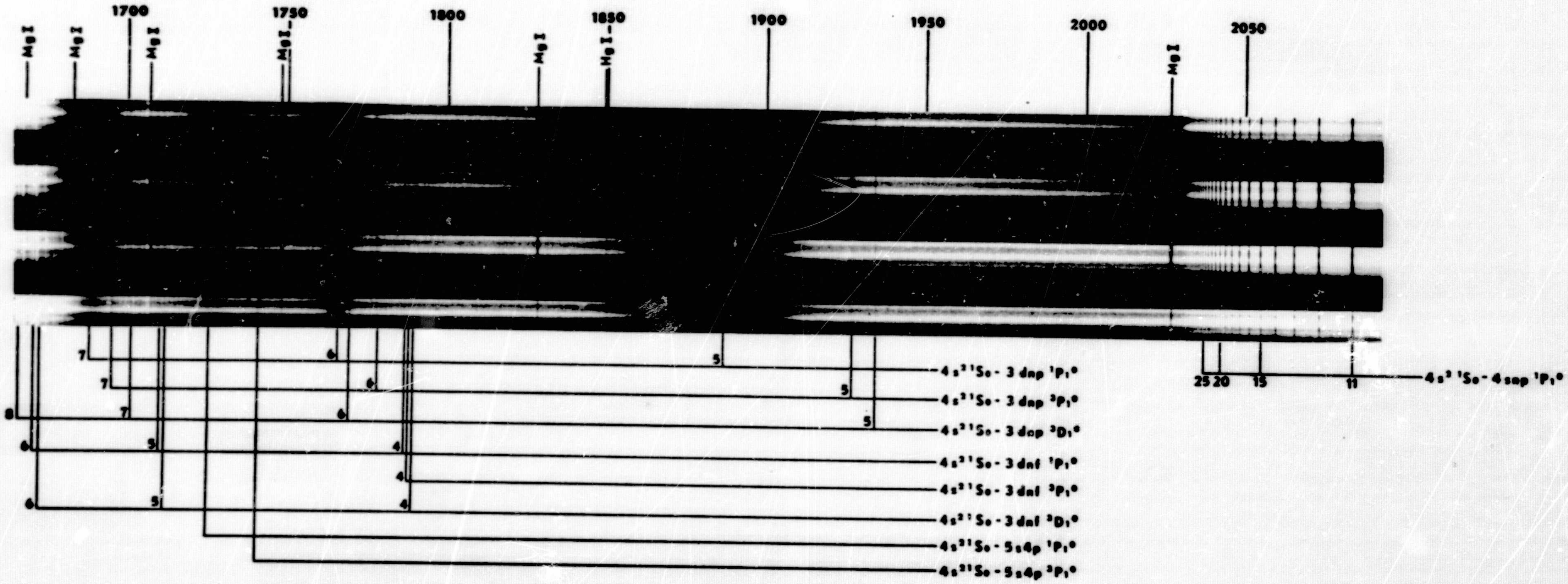


Plate 1. Calcium vapor absorption spectra.

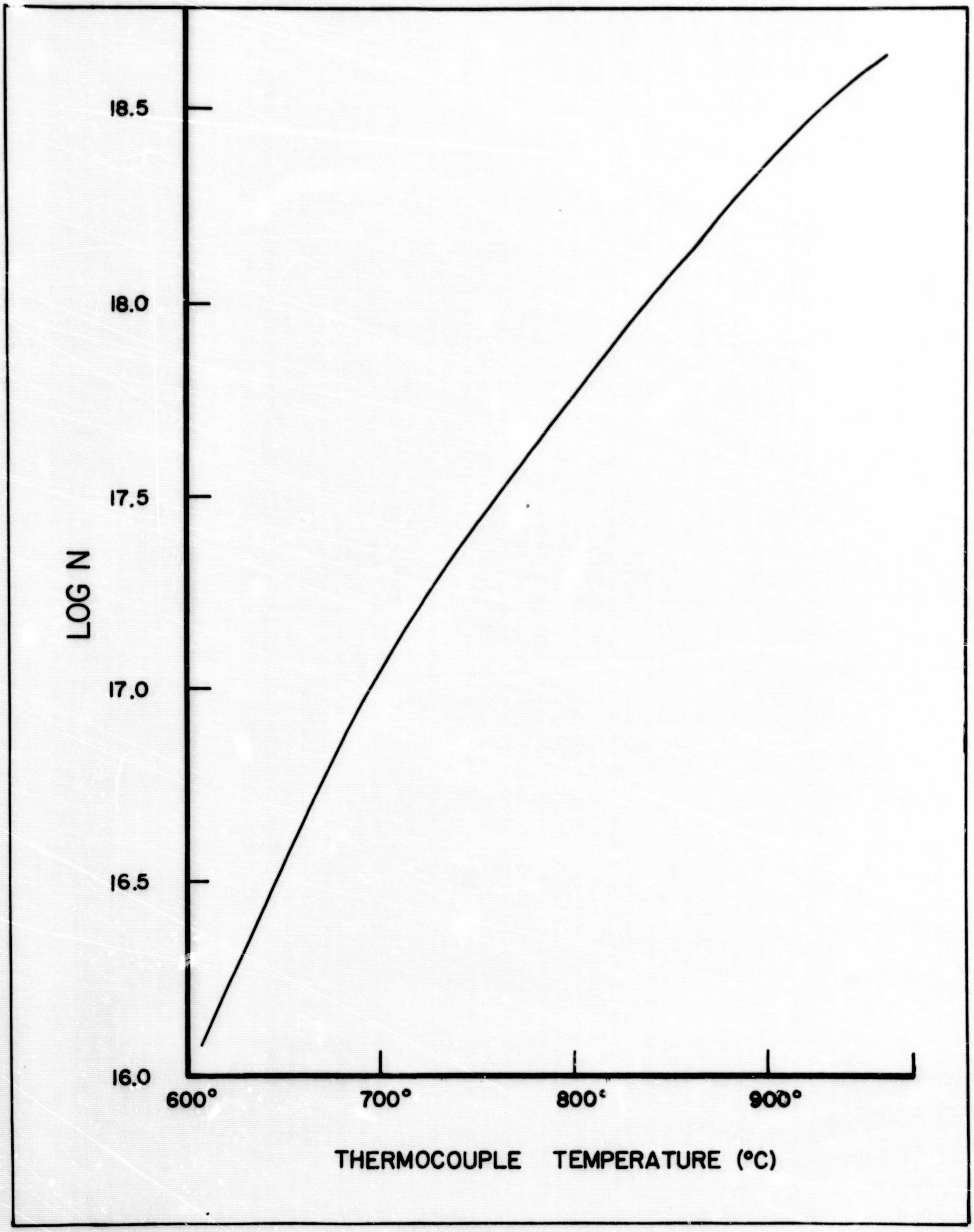


Figure 1. Logarithm of the number of absorbing atoms in the line of sight as a function of thermocouple temperature.

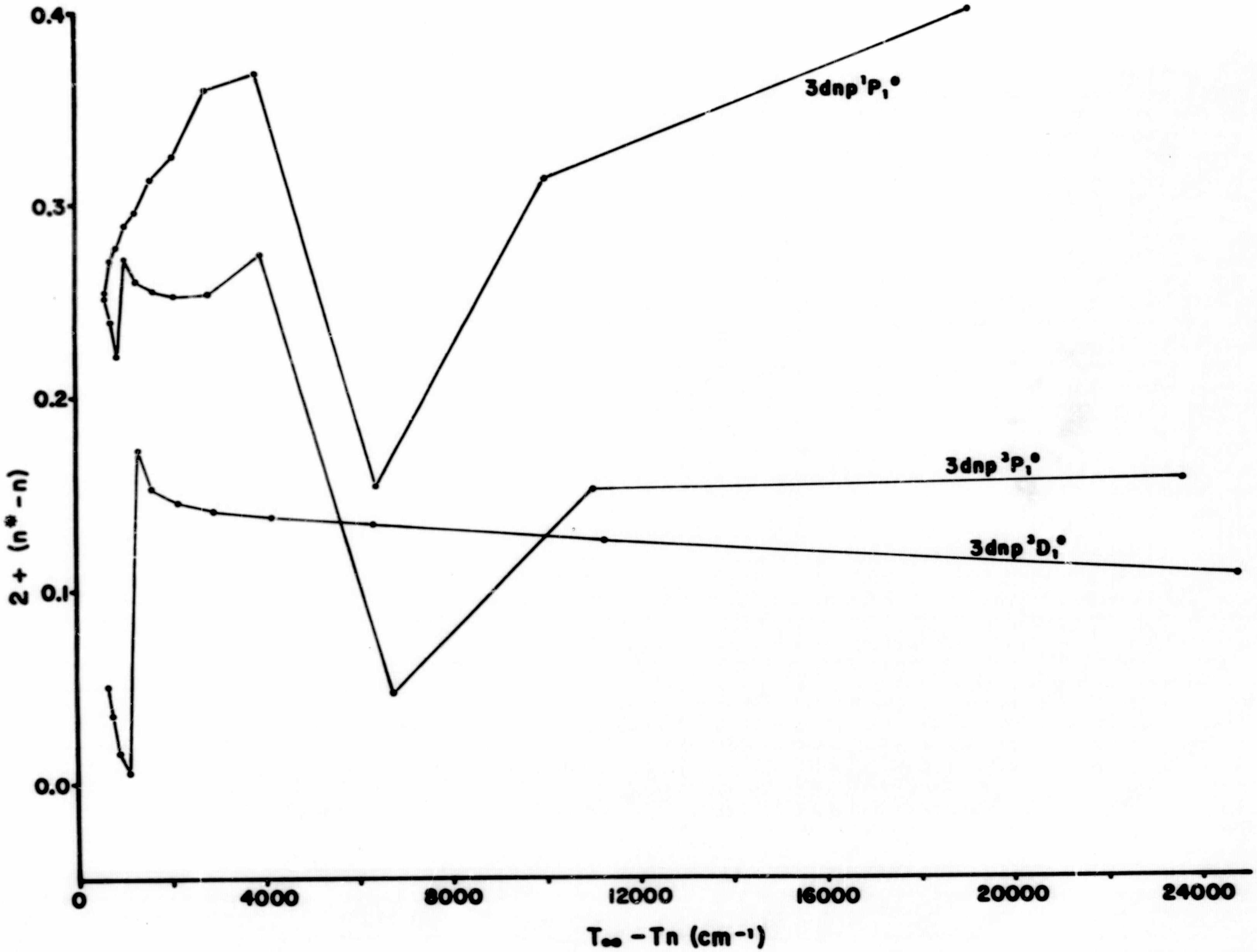
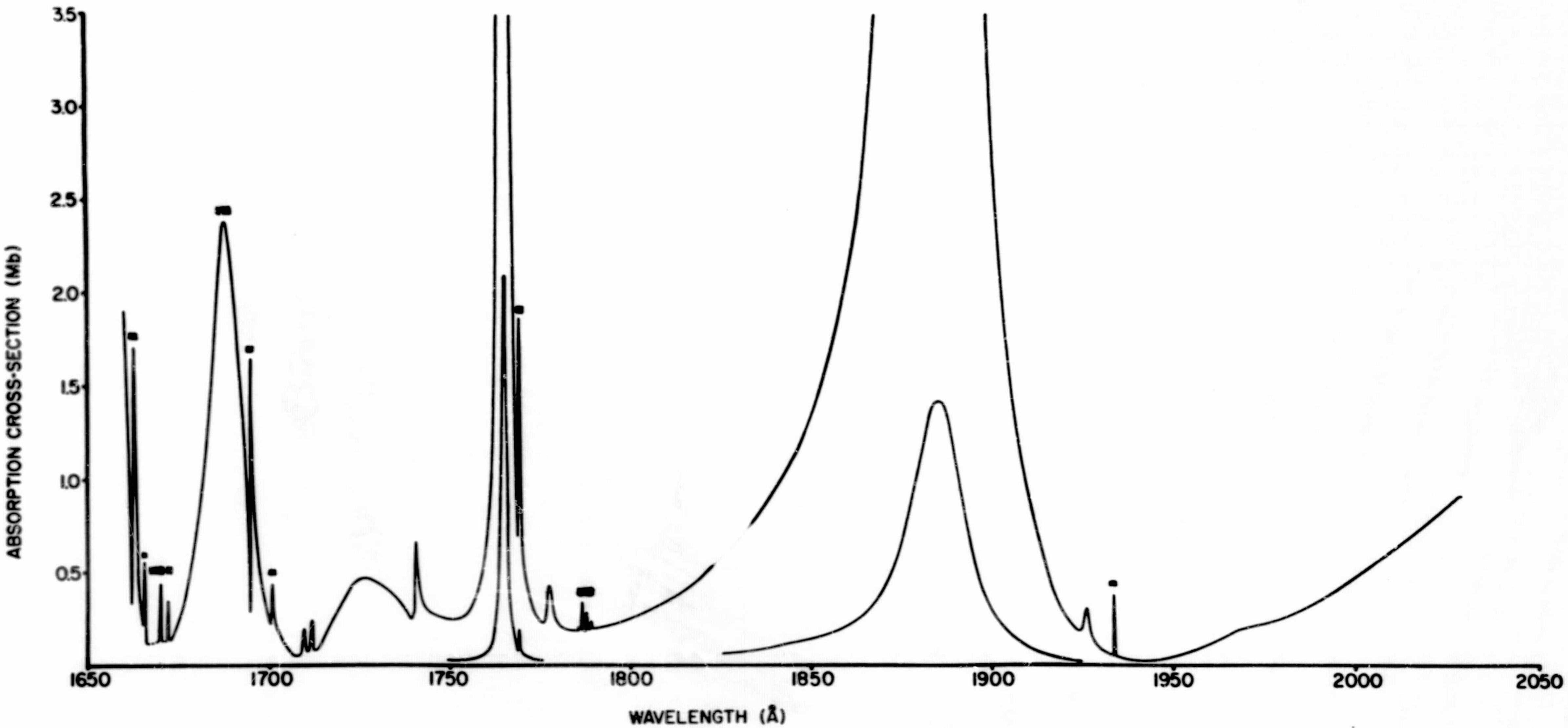


Figure 3. Variation in quantum defect for the 3dnp series in calcium.



ABSORPTION CROSS-SECTION OF CALCIUM VAPOUR

Figure 2. The atomic absorption cross-section as a function of wavelength. Wavelength regions marked with a dark line were not adequately resolved by the monochromator or were not adequately observed to yield accurate cross-sections. The lines at 1885.9 and 1765.1 Å are also shown reduced by a factor of ten.

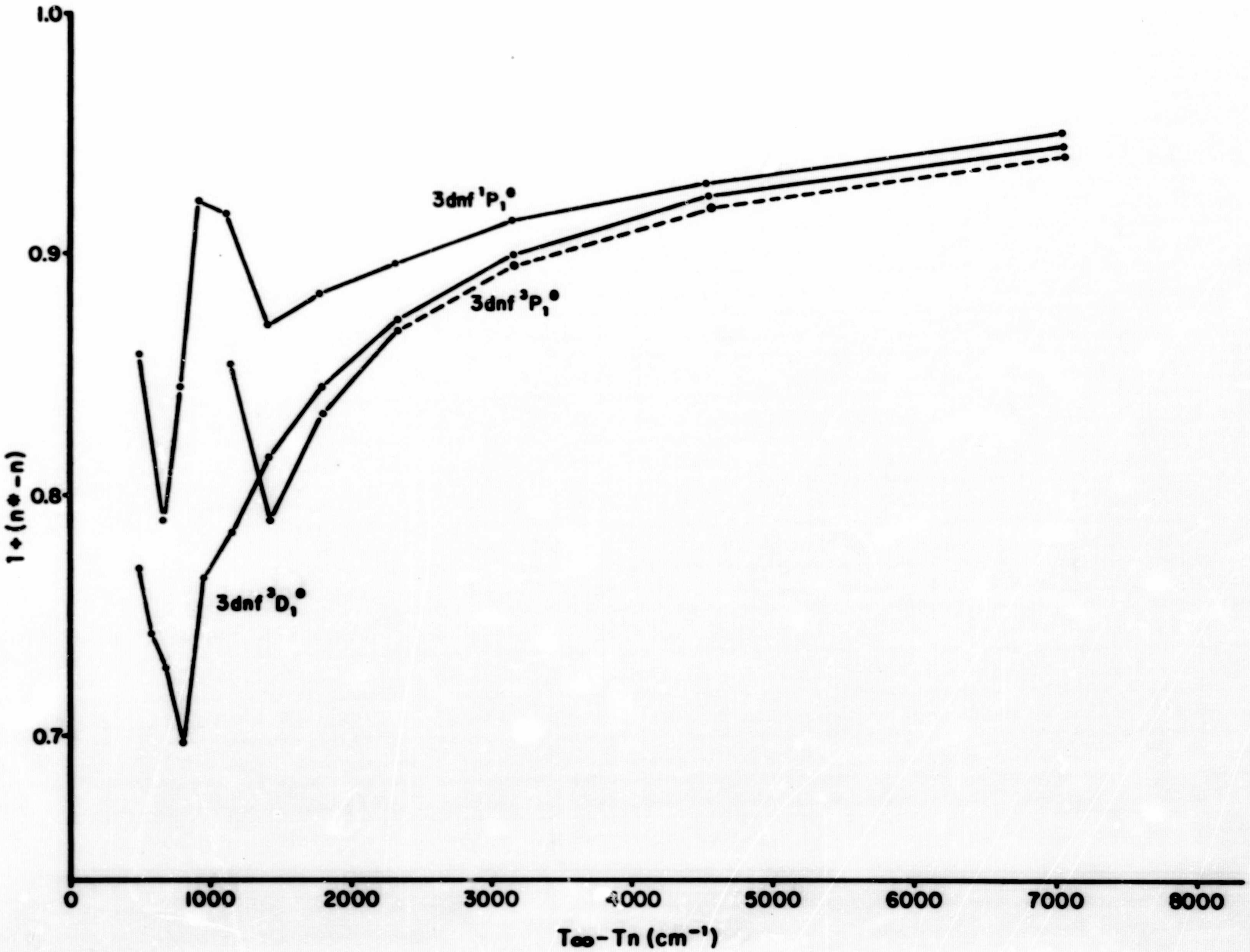


Figure 4. Variation in quantum defect for the 3dnf series in calcium. The open circles represent unobserved levels.

Table 1

Trace	Temp. range	Scan speed (Å/min)	Scan direction	Wavelength range
22	545-595	200	Violet	1650-2250
23	625-645	50	Violet	1650-2250
24	840-845	50	Violet	1650-2230
25	844-855	20;50	Red	1650-2150
26	898-905	50	Violet	1650-2150
27	928-939	50	Red	1650-2250
28	740-762	50	Violet	1828-2135
29	668-683	100	Violet	1675-2150
30	603-638	100	Violet	1675-2150
31	Calibration of electrometer gain ratios			
32	520-560	50	Violet	1675-2120
33	500-515	100	Red	1675-2130

Table 2

Ca I $4s^2\ ^1S_0 - 3dnp\ ^1P_1^\circ$

Limit: 63017.0 cm^{-1}

n	λ(Å)	$\upsilon(cm^{-1})$	f-value	n*	Half-width cm^{-1}
5	1885.9	53025.	1.17×10^{-2}	3.314	507
6	1765.1	56654.	1.72×10^{-3}	4.153	51
		$4s^2\ ^1S_0 - 3dnp\ ^3P_1^\circ$			
		Limit: 62956.3 cm^{-1}			
5	1926.5	51908.	1.25×10^{-5}	3.151	46
6	1777.6	56254.	1.48×10^{-5}	4.046	47
		$4s^2\ ^1S_0 - 3dnp\ ^3D_1^\circ$			
		Limit: 62956.3 cm^{-1}			
5	1933.88	51709.5	- - -	3.124	
6	1768.92	56531.7	- - -	4.133	
		$4s^2\ ^1S_0 - 3dnf\ ^1P_1^\circ$			
		Limit: 63017.0 cm^{-1}			
4	1786.28	55982.3	- - -	3.950	
5	1709.4	58500.	5.1×10^{-6}	4.929	27
6	1670.01	59879.9	- - -	5.914	
		$4s^2\ ^1S_0 - 3dnf\ ^3P_1^\circ$			
		Limit: 63017.0 cm^{-1}			
4	1787.42	55946.6	- - -	3.940	
		$4s^2\ ^1S_0 - 3dnf\ ^3D_1^\circ$			
		Limit: 62956.3 cm^{-1}			
4	1788.82	55902.8	- - -	3.944	
5	1711.5	58428.	6.0×10^{-6}	4.923	20
6	1672.15	59803.2	- - -	5.899	

$4s^2\ ^1S_0 - 5s4p\ ^1P_1^\circ$ $\lambda = 1724.8$ $\upsilon = 57978\ cm^{-1}$

$4s^2\ ^1S_0 - 5s4p\ ^3P_1^\circ$ $\lambda = 1740.2$ $\upsilon = 57465\ cm^{-1}$ $f = 3.0 \times 10^{-5}$

Half-width = 33 cm^{-1}

Table 3

Possible new autoionizing lines seen in the shock tube.

λ_{air}(Å)	Possible indentification
5742	4s6s $^1S_0$ - 3d7p $^1P^\circ_1$
4703	4s4d $^3D_3$ - 3d7p $^3P^\circ_2$
3856	4s5s $^3S_1$ - 4p5s $^3P^\circ_2$ or 4s5s $^1S_0$ - 3d7p $^1P^\circ_1$
3170	4s3d $^3D_3$ - 3d5p $^3P^\circ_2$
2929	4s3d $^1D_2$ - 3d4f $^1P^\circ_1$
2786	4s3d $^3D_3$ - 3d6p $^3P^\circ_2$
2761	4s3d $^3D_3$ - 3d6p $^3D^\circ_3$

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13. ABSTRACT

The vacuum ultraviolet absorption spectrum of calcium vapor was observed in an attempt to discover transitions to new energy levels. Seven new levels have been found between 51000 and 59000 cm^{-1}, three of which are ascribed to the 3d5p, 3d6p, and 5s4p configurations, with the remaining four lines resulting from transitions to 3d4f and 3d5f configurations. The 3d5p $^3P^\circ_1$ level reported by Ditchburn and Hudson at 52225 cm^{-1} is now found at 51909 cm^{-1}. A graph of relative cross-section for photoionization from the ground state of calcium is shown between 1660 Å and the principal series limit. A shock tube was used to observe several transitions to auto-ionizing levels from excited states.

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